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WATER CONTAMINATION IN FALLOUT AREAS

ROBERT A. TAFT SANITARY ENGINEERING
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Robert A. Taft Sanitary Engineering Center

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WATER CONTAMINATION IN FALLOUT AREAS

INTRODUCTION

The objectives of this project were to evaluate the radiological hazard in Air Force water supplies contaminated by fallout, and to develop design criteria for protective devices.

A review of classified and unclassified literature was made to obtain present thinking and knowledge about maximum permissible concentrations (MPC) in water under emergency conditions and the nature and behavior of fallout in surface waters. Where gaps existed in information, field and laboratory studies were made so that subsequent design and cost evaluation of various treatment devices would be meaningful.

LITERATURE REVIEW

HAZARD

An inspection of Air Force records indicates 43 percent of the water supplies of Air Force bases in continental United States come from Air Force owned wells, 54 percent from municipally owned wells and surface sources, and 3 percent from Air Force owned surface supplies. (See Appendix A). In addition a majority have substantial underground storage of finished water. This situation reduces the magnitude of the problem since ground water supplies should be safe from fallout contamination and only the surface supplies will need some supplemental treatment following contamination by fallout from high yield thermonuclear weapons.

Present information is inadequate for an evaluation of all the pertinent factors affecting the short-term hazard of water contamination by fallout. However, it is apparent that water contamination is not likely

to significantly reduce the military effectiveness of personnel using it except where the general level of contamination is extremely heavy (greater than 1,000 r/hr at H + 1). Under such circumstances the external radiation hazard will be a major cause of casualties, and water contamination will be of relative concern only where shelter is provided to reduce external radiation exposure. Shelter also will be necessary to detect dangerous radiation levels in raw or finished water.

The uncertain basis for the selection of emergency MPC values makes the matter of detection and degree of treatment correspondingly uncertain. For instance, calculations of the radiation dose to various organs due to ingestion of water contaminated by fresh mixed fission products indicate that the gastro-intestinal tract is the most heavily irradiated organ of the body under conditions of short time ingestion. On the other hand, many radiation biologists consider that the greatest hazard from ingestion of mixed fission product radioactivity is due to the increase in body burden of Sr^{90} and that radiation damage to other organs and by other nuclides is of secondary importance.

This latter concept means that during the period immediately after fallout occurs, when external radiation levels are highest, the hazard from ingestion of a unit of gross fission product activity is less than at a later time when the percentage of the activity present due to Sr^{90} is greater.

Table I shows computations of the levels of activity of ideal fission product mixtures of various ages that would lead to a body burden of 5 μc of Sr^{90} if used for one year together with estimates of radiation doses to the gut and to the skeleton due to use of such water.

TABLE I

GROSS FISSION PRODUCT ACTIVITY (μc/ml) AND DOSE (rep)
TO GUT AND SKELETON AT VARIOUS TIMES

Age at Start of Use	Gross Fission Product Activity for 365 Day Consumption*	Total Dose to Gut** for 365 Day Consumption (All Fission Products)	Dose to Skeleton*** For 365 Day Consumption	
			Dose During 1st Yr.	Dose to Infinity
1 Day	1.1 μc/ml	750 rep	195 rep	675 rep
10 Days	7.0×10^{-2} μc/ml	500 rep	165 rep	650 rep
30 Days	2.0×10^{-2} μc/ml	335 rep	125 rep	600 rep
90 Days	5.0×10^{-3} μc/ml	185 rep	55 rep	550 rep
365 Days	9.5×10^{-4} μc/ml	70 rep	5 rep	500 rep

* Consumption for 365 days will accumulate a Sr^{90} body burden of 5.0 μc.

** Dose from all fission products to large bowel (assumes no G.I. Absorption)

*** Dose from Sr^{89}, Sr^{90}-Y^{90}, Sr^{91}-Y^{91}, Zr^{95}-Nb^{95}, Ba^{140}-La^{140}, and rare earths.

From these idealized considerations it would appear that maximum permissible concentrations of radioactivity in drinking water under military emergency conditions might well be in the order of 0.1 μc/ml during the early period after attack and that at later times much lower levels would have to be maintained. (Appendix B gives a few details on instrumentation under such circumstances, and also some of the limitations).

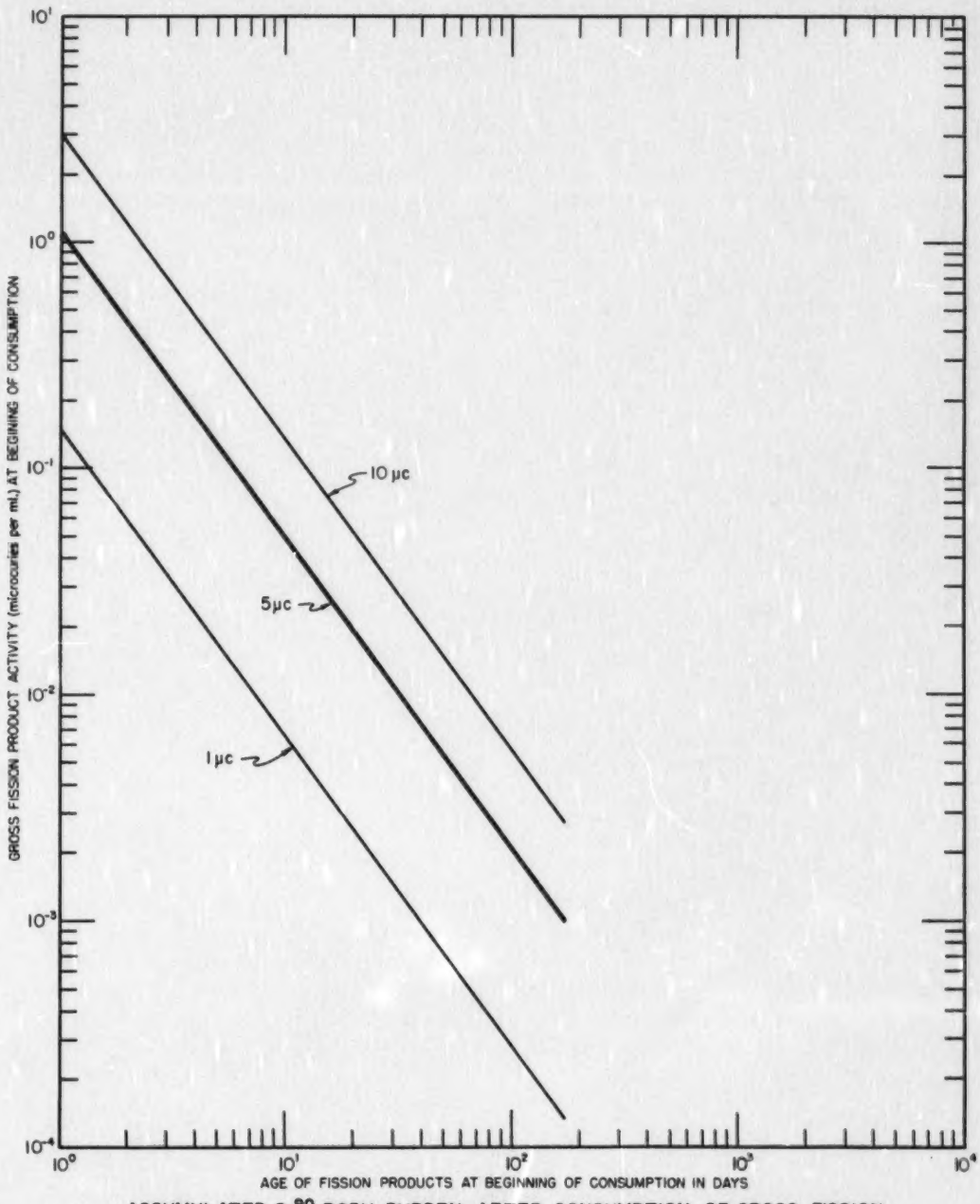
However, it is understood that the nature of the combat situation will many times determine the time of consumption and level of activity to endure. Figure 1 illustrates what gross activity level will result in a Sr^{90} body burden of 1, 5 and 10 μc after daily consumption for one year, and would thus aid in making command decisions.

Any strontium fractionation during dissolving or passage through treatment process would cause these curves to be lowered in proportion to the percentage increase in strontium.

Alpha contamination has not been considered to be a serious fallout problem since Pu^{239} is easily removed by normal water treatment processes and by adsorption on soil particles and other surfaces.

EFFECTIVENESS OF CONVENTIONAL WATER TREATMENT

Conventional water treatment processes remove only 10 to 70 percent of those soluble radionuclides which present the greatest hazard when ingested. Particulates, however, are very well removed. Supplemental treatment with distillation or ion-exchange equipment will reduce the gross activity in solution by 99 to 99.9 percent. Since most of the past water decontamination studies have been made with simulated reactor or fuel processing wastes, this project was started to determine if actual primary


GROSS FISSION PRODUCT ACTIVITY (microcuries per ml.) AT BEGINNING OF CONSUMPTION
AGE OF FISSION PRODUCTS AT BEGINNING OF CONSUMPTION IN DAYS
10 μc
5μc
1μc

ACCUMULATED Sr^{90} BODY BURDEN AFTER CONSUMPTION OF GROSS FISSION PRODUCTS IN DRINKING WATER FOR 1 YEAR

Figure 1

fallout would have similar nuclear properties and similar response to treatment as reactor wastes.

NEVADA FALLOUT STUDIES

SAMPLE COLLECTION

Dust samples were collected in fallout trays placed at various distances from the ground zero of the "Priscilla" balloon shot, detonated June 24, 1957. A few days later soil samples were vacuumed from the desert floor at about 30 yards from ground zero. These samples were then sized into the three groups of <5, 5 to 50, and >50 microns, and shipped to this Center for solubility and radiochemical tests.

The sample collection was done jointly with representatives of the Engineer Research and Development Laboratory, U. S. Army.

LABORATORY TEST PROCEDURES

A gross beta activity determination was made on each sample by counting the radioactivity in a raw (unaltered) soil portion with a GM counter, and also in an acid-dissolved portion with an internal, gas-flow, proportional counter. By repeating this counting procedure at frequent intervals over a 6-month period, it was possible to establish the rate of decay for any sample. (Appendix C describes the counting techniques and instruments used).

Another portion of each soil sample was mixed with 10 liters of distilled water. A small amount of 1 $\underline{N}$ HNO_3 was added to keep the pH about 7.

To obtain a representative sample of this slurry for assay, a rapid mixer was used for a few minutes before and during the siphoning of two 100-ml portions. These duplicate samples were each put through a membrane filter to separate the suspended solids from the dissolved solids. Both portions were heated to dryness, plated, and assayed for gross beta activity with an internal proportional counter. The same sampling technique was used to obtain larger portions for subsequent radiochemical analysis.

SOLUBILITY OF FALLOUT

As indicated in Table II, about 10 percent of the radioactive material in each particle size range usually went into solution. However, the fine dust (<5 microns) at approximately 4000 yards from ground zero was only 3 percent soluble. There is no apparent reason for this low value, though it should be noted that this dust had 10 to 30 times the specific activity of the other soil fractions.

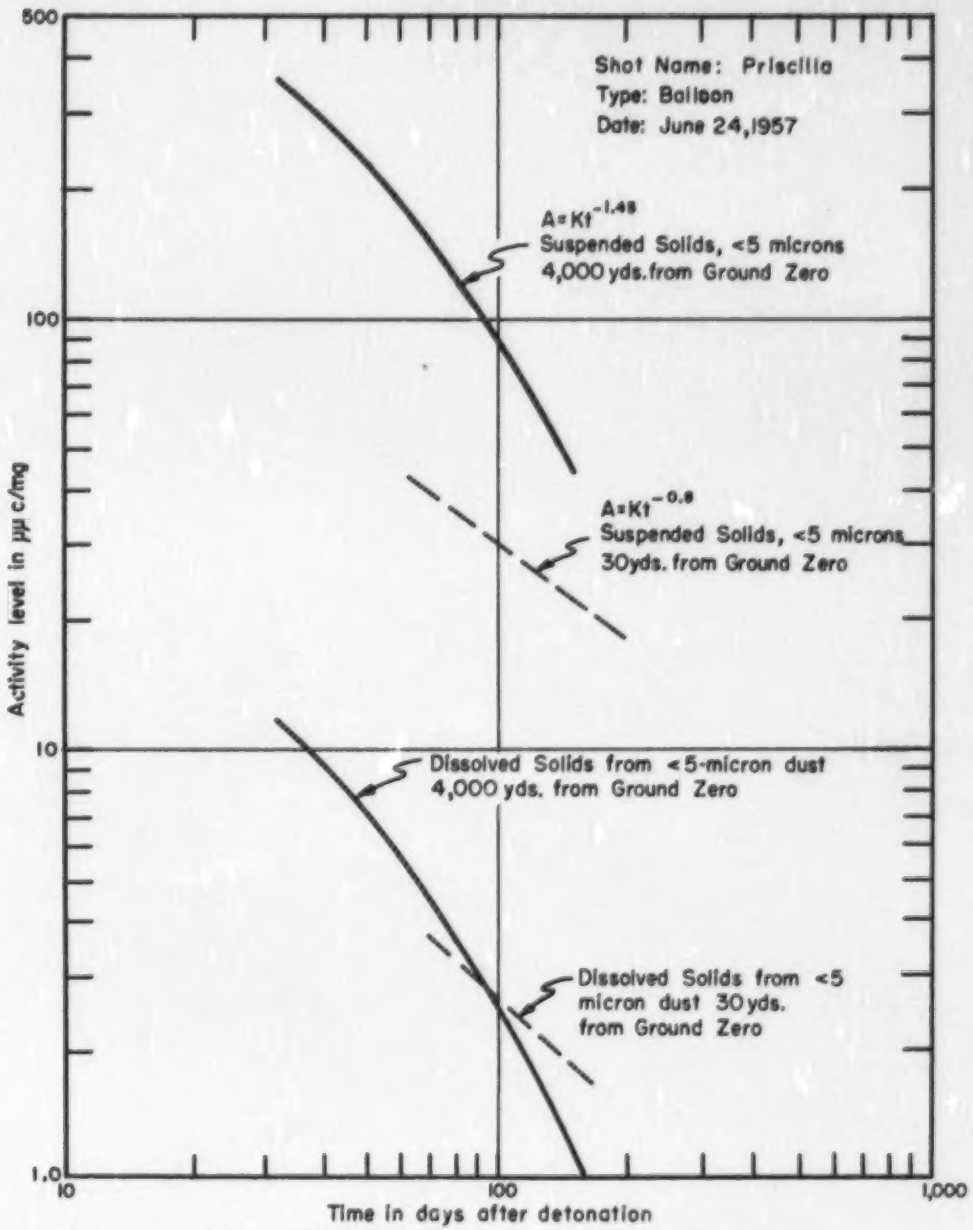
The percent of radioactive soluble material in each case did not change significantly over a 4 to 5 month period. The decay rate for the suspended solids was similar to the rate for the soluble portion. (Figure 2).

Figure 2 illustrates the decay rate for the near ground zero soil to be somewhat lower than for the samples collected at 4000 yards from ground zero. The other two fractions of soil in each case had similar decay rates, so these curves were not plotted on this figure. Subsequent radiochemical work showed a foreign radionuclide to be present only in the near ground zero sample, so this may explain the distinct difference in the decay rates.

TABLE II

SOLUBILITY OF PRIMARY FALLOUT
NEVADA TEST, JUNE 24, 1957

Sample Size in microns	Source	Date Counted	Gross Beta Radioactivity in μμc/mg of Fallout Particulates Added: Suspended Solids	Dissolved Solids	Total	Percent DS/Total
<5	30 yds.	8/24/57	44.	4.9	48.9	10.
	from	10/24/57	24.	2.6	26.6	9.8
	Gr.Zero	12/24/57	18.	1.8	19.8	9.1
5-50	30 yds.	8/24/57	39.	3.7	42.7	8.7
	from	10/24/57	24.	2.2	26.2	8.4
	Gr.Zero	12/24/57	16.5	1.5	18.0	8.3
>50	30 yds.	8/24/57	40.	4.4	44.4	9.9
	from	10/24/57	23.	2.3	25.3	9.1
	Gr.Zero	12/24/57	17.	1.6	18.6	8.6
<5	4000 yds.	7/24/57	400.	14.	414.	3.4
	from	8/24/57	170.	6.	176.	3.5
	Gr.Zero	10/24/57	69.	1.5	70.5	2.2
		11/24/57	48.	0.9	48.9	1.9
5-50	4000 yds.	7/24/57	23.	4.0	27.0	12.
	from	8/24/57	10.	1.7	11.7	14.
	Gr.Zero	10/24/57	4.2	0.56	4.76	12.
		11/24/57	3.0	0.36	3.36	11.
>50	4000 yds.	7/24/57	11.	1.6	12.6	13.
	from	8/24/57	5.	0.75	5.75	13.
	Gr.Zero	10/24/57	2.1	0.30	2.40	12.
		11/24/57	1.5	0.20	1.70	12.



DECAY CURVES FOR PRIMARY FALLOUT MIXED IN WATER

Figure 2

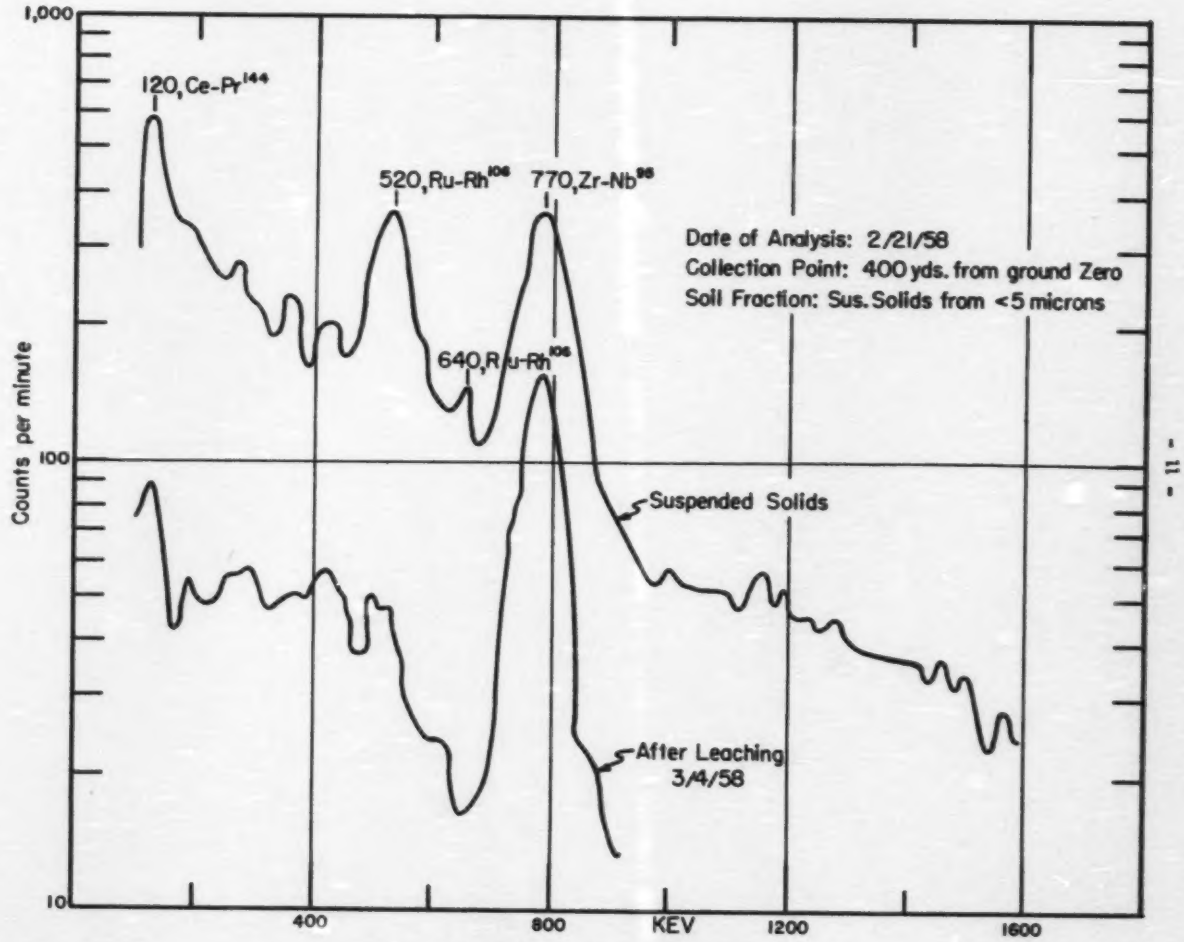
Previous studies of the solubility of dry fallout particulates have generally shown somewhat lower solubility than was found in this study although the data available are notably variable. No rational explanation is available for the wide variations that have been found in various studies.

It is obvious that there are major differences in solubility between fallout from detonations over land and those over water. For practical purposes in estimating water contamination from detonations over deep water, one should consider essentially all of the activity to be soluble. For detonations over land a solubility of about 10 percent seems to be a reasonably conservative figure for estimates of hazard.

RADIOCHEMICAL ANALYSES OF SOIL AND WATER

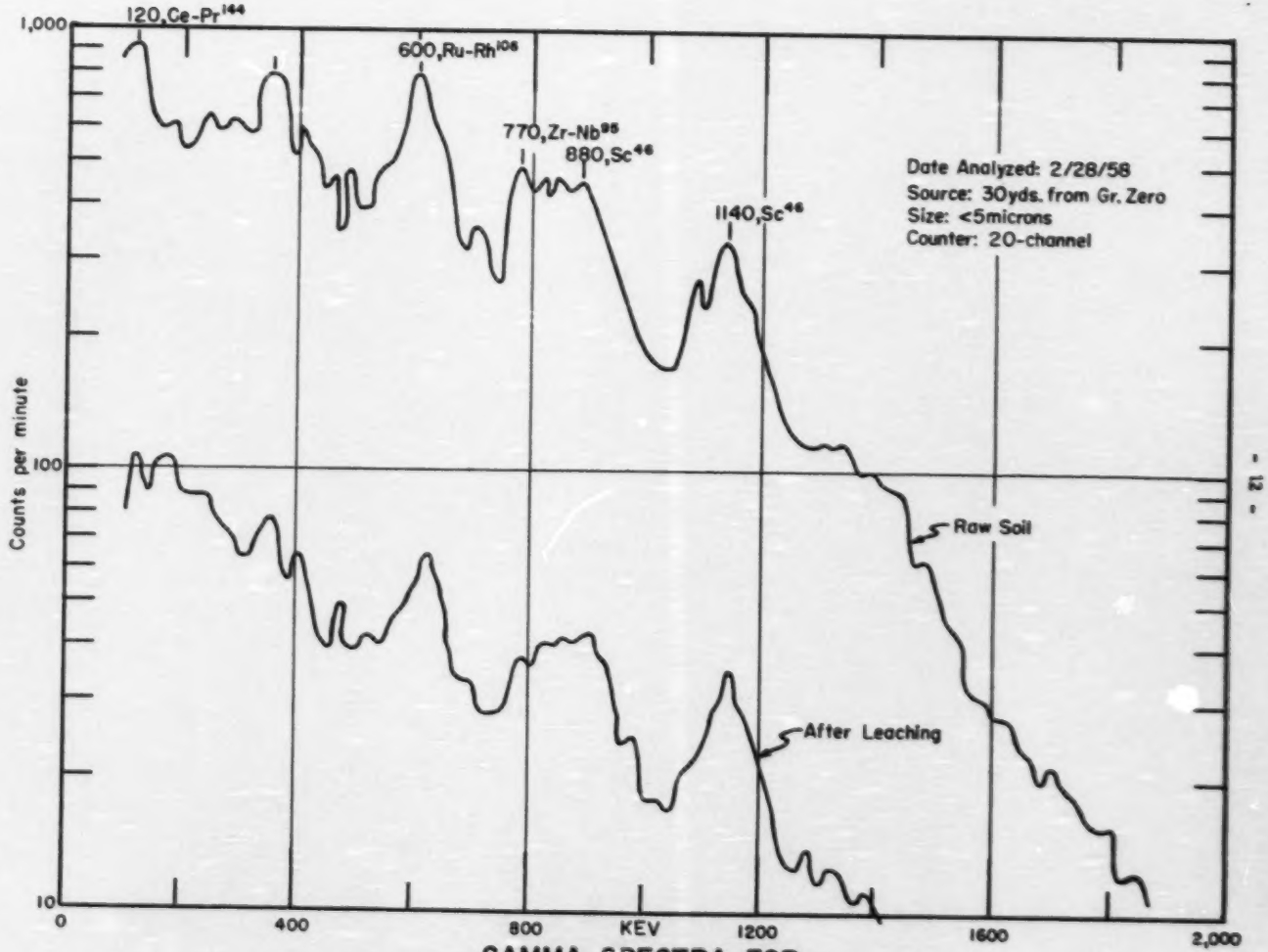
A complete analytical procedure for strontium is given in Appendix D. Considerable trouble was encountered with the analysis for strontium in the Nevada soil and many modifications of the usual procedures were tried to improve reproducibility of the results. The high calcium content of the soil made it desirable to not dissolve the sample completely by using HF, $HClO_4$ and HCl since this complicated the subsequent steps in separation. The method finally adopted used hot 6M HNO_3 which removes only about 75 to 80 percent of the total activity, but permits a much better and consistent strontium recovery from the leachate.

Figures 3 and 4 are gamma spectra at D + 190 for two raw soil samples and also for the acid leach of each. It is apparent that Ce^{144}-Pr^{144}, Zr^{95}-Nb^{95}, Ru^{106}-Rh^{106}, are present in both samples. An additional activity



GAMMA SPECTRA FOR
SOIL MIXED IN WATER AND ACID-LEACHED

Figure 3



GAMMA SPECTRA FOR
FALLOUT MIXED WITH WATER AND ACID LEACH

Figure 4

with gamma rays characteristic of Scandium^{46} is found in the sample collected 30 yards from ground zero. This is not a fission product and would not be expected in fallout.

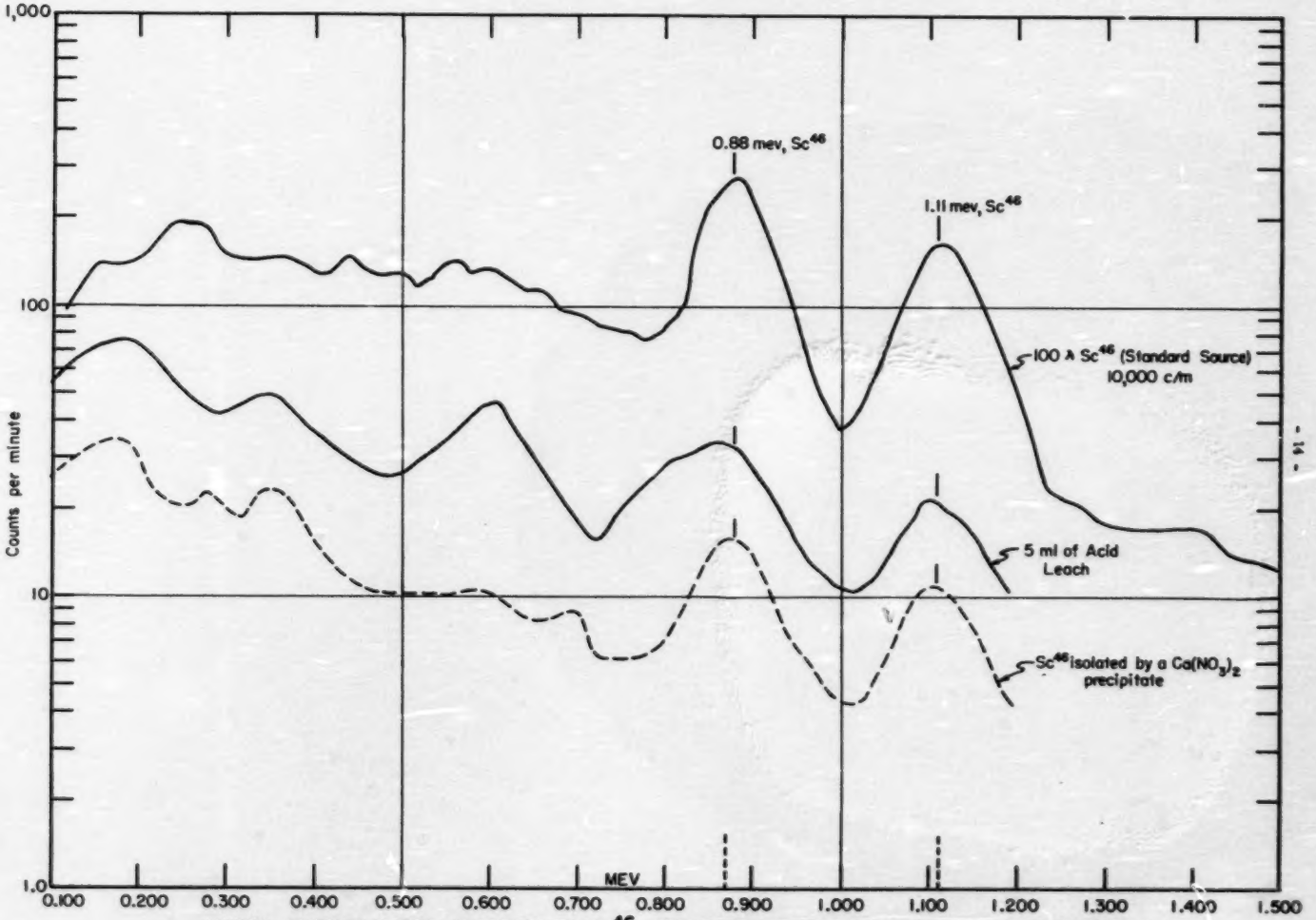
A chemical separation was attempted to determine whether this activity was due to Sc^{46}. Figure 5 shows the gamma spectra for the acid-leached soil from near ground zero and also the chemically isolated Sc^{46} with peaks at 0.88 and 1.12 mev. The upper curve is the gamma spectrum of a 100 microliters Sc^{46} standard ($\sim$10,000 cpm).

It is odd that Sc^{46} should show up in all three fractions of soil collected near ground zero, but not in samples of dust from 4000 yards away.

Radiochemical determinations made at D + 30 indicated the I^{131} levels to be about 1 $\mu\mu$c/mg for all fractions from 4000 yards except the <5 micron fraction which was approximately 10 to 15 times higher.

Strontium levels in the soil were low and difficult to determine because of the high calcium content. Some of the data from the near ground zero samples in Table III indicate that ten months after the detonation about one percent of the total beta activity in the soil was due to strontium.

The determination of the degree of solubility of strontium as compared to the overall fission product solubility was one of the main objectives of this study. Table III illustrates that strontium is preferentially dissolved by a factor of approximately 5. This is not a large fractionation but it can be considered significant and should alter the maximum permissible gross activity level correspondingly.



GAMMA SPECTRA FOR ACTIVITY IN Sc^{46} STANDARD, ACID SOIL LEACH, AND CHEMICALLY ISOLATED Sc^{46} FROM SOIL COLLECTED 30 YDS. FROM GROUND ZERO

Figure 5

TABLE III

RELATIVE SOLUBILITY OF STRONTIUM IN WATER
NEVADA TEST SHOT, JUNE 24, 1957

Sample									
Size Microns	Source	Date	Gross Beta in Soil μμc/mg	Strontium in soil μμc/mg	$\text{Ratio}_1 = \frac{\text{G.B}_{soil}}{\text{Sr}_{soil}}$	Gross Beta in Soluble Portion μμc/l	Strontium in Soluble Portion μμc/l	$\text{Ratio}_2 = \frac{\text{G.B}_{water}}{\text{Sr}_{Water}}$	$\text{Ratio}_3 = \frac{\text{Ratio}_1}{\text{Ratio}_2}$
<5	30 yds. from Gr.Zero	4/21/58	22	0.11	200	2500	68	37	5.4
5-50	30 yds. from Gr.Zero	4/21/58	22	0.14	160	2100	70	30	5.3
>50	30 yds. from Gr.Zero	4/21/58	19	0.18	110	1700	73	23	4.8

DECONTAMINATION BY NATURAL AND CONVENTIONAL TREATMENT

Plain Settling

Table IV indicates that where settling takes place for 16 to 24 hours, there will be a 62 to 92 percent removal of the radioactive material. Hence, an open reservoir will serve as a good protective device in removing particulate matter.

Coagulation

Supernates from the overnight settling process were coagulated with 20 mg/l of ferric chloride plus lime when necessary to adjust the pH. Removals varied from 4 to 76 percent. Based on previous experience with radioactivity in ionic form, this was to be expected.

Filtration

By using a membrane filter, it was possible to remove all suspended material from a mixture of fallout and water. A conventional sand filter would be somewhat less effective so filter plants could not hope to remove much more than 85 to 90 percent of the activity.

Combined Treatment

Combined treatment will remove most of the radioactive particles, but only about 10 to 70 percent of the dissolved radionuclides. Ion-exchange resins can, however, be used to reduce the dissolved activity by 99 + percent. Since extensive research had been done previously on the efficacy of ion-exchange resins for removing radioactivity, no further tests were made during this project.

TABLE IV

REMOVAL OF RADIONUCLIDES FROM MIXTURE OF FALLOUT AND WATER
BY SETTLING, COAGULATION AND FILTRATION
NEVADA TEST SHOT, JUNE 24, 1957

Particle size Microns	Source	Date Counted	Gross Beta Radio-activity μμc/l	Plain Sedimentation		Coagulation of Settled Water with 20 mg/l of $FeCl_3$					Filtration of Raw Mixture by Membrane Filter	
				Gross Beta Remaining μμc/l	Percent Removal	Gross Beta Remaining μμc/l	Initial pH	Lime added mg/liter	Final pH	Percent Removal	Gross Beta Remaining μμc/l	Percent Removal
<5	30 yds. from Gr.Zero	9/24/57	71,000	27,000	62	23,000	8.3	-	7.1	15	7,000	90
5-50	30 yds. from Gr.Zero	9/24/57	79,500	24,500	69	23,600	8.3	-	7.0	4	7,000	91
>50	30 yds. from Gr.Zero	9/24/57	67,000	33,000	64	28,000	8.4	-	7.3	15	6,100	89
<5	4000 yds. from Gr.Zero	8/20/57	31,000	2,600	92	900	6.7	26	6.8	65	970	97
		2/24/58	4,000	(Re-run on remaining portion of sample)							110	97
5-50	4000 yds. from Gr.Zero	8/20/57	15,000	2,400	84	1,500	7.2	7	7.2	38	1,900	87
		2/24/58	2,400	(Re-run on remaining portion of sample)							200	92
>50	4000 yds. from Gr.Zero	8/20/57	14,000	3,800	73	900	7.8	7	7.8	76	2,000	86
		2/24/58	2,700	(Re-run on remaining portion of sample)							140	94

DESIGN CRITERIA

Where only surface water is available, conventional water treatment and distribution systems must suffice for large requirements such as fire protection and radioactive surface decontamination. Ion-exchange and distillation are not economically feasible to prepare water for such uses.

It is anticipated that for the first 10 days after fallout, a supply of one gallon per person per day will suffice for drinking and culinary purposes. This figure represents roughly 1/100 of the average water demand. For an installation with an average water use of 100,000 gallons per day, it will be necessary then to supply 1,000 gallons per day that is safe for drinking and cooking. Ion-exchange treatment is the most practical and economical method of supplying this water.

Fresh fission products contain activity due to anions and cations so that a mixed bed ion-exchange column will be required. The anions are mostly short-lived nuclides and a cation bed alone will suffice for the long-term requirements.

TYPE, SIZE AND COST OF UNITS

For Immediate Demand

Although a centrally located and operated demineralizer could produce water more economically than smaller more widely dispersed units, they have not been considered as the most desirable solution because of the problem of distributing the water. At times when such treatment would be needed, it would be most important to avoid all unnecessary exposure to external radiation. Distribution of a centrally treated supply for

drinking and culinary purposes by tank truck or other means would involve considerable radiation exposure to personnel involved which could be avoided by providing numerous small demineralizers in shelters or at other points of water use for decontaminating the piped water supply.

Small, mixed-bed demineralizers similar to those in common use in laboratories would be equipped with disposable cartridges containing the ion-exchange resins. A simple conductivity meter or resin color change would indicate the need for replacement of an exhausted cartridge. Under emergency conditions, it will be more practical to discard the exhausted cartridges than to regenerate ion-exchange resins. This type of apparatus is recommended to supply the immediate requirements for all installations.

The typical cost estimates presented in Table V for small demineralizers are based upon the following conditions:

1. Total solids content of water to be treated is 10 grains per gallon which is an average quality (1 grain per gallon = 17.1 mg/l).
2. Flow rate of 5 gallons per hour (2 gpm/cu. ft.).
3. Each demineralizer cartridge has a capacity for 1100 grains of dissolved solids--thus 110 gallons/cartridge; or 9 units needed per 1000 gallon per day.
4. Maximum cartridge change of one per day.

TABLE V

COST OF DEMINERALIZING RADIOACTIVE WATER
FOR SHORT-TERM

Water Demand gpd	Complete Units Needed	Initial Cost of Complete Units at $81. ea.	Operating (1) Cost Based On Cartridge Needs
10,000	90	$ 7300	$ 870
5,000	45	3650	435
1,000	9	730	87 (9 cents/gal)

(1) Based on average water quality. Where the total solids in the water are less, the operating cost will be less.

It is possible to use a much simpler throw-away ion-exchange cartridge and holder, providing it is not put into a pressure line. The capacity for such a model is 900 grains with a rate of 5 gph. The initial wall bracket support, faucet, and hose connections would cost about $7.50 each and the replaceable cartridge about $9.50. Prices would vary with size of order, but a 30 percent reduction is possible. This would make the initial cost 10 times less than the other example of costs, but the operating cost would be about the same.

Long-term Demand

A pressure cation-exchange bed operated on the sodium cycle is the preferred method of decontaminating water for drinking and cooking purposes where this is necessary on a long term basis. This equipment would normally be located at the water treatment plant and operated by

that plant's personnel. Provisions for distribution of the water with proper sanitary precautions would need to be taken.

The cost estimates presented in Table VI for such units are based upon the following conditions:

1. Dissolved solids content of water to be treated is 8 gr/gallon.
2. Maximum flow rate of 8 gpm/sq.ft.
3. All water will be treated.
4. A high capacity styrene resin will be used, such as Dowex-50 or Amberlite IR-120.
5. Capacity of resin is 30 kilograins per cu.ft.
6. Amount of regenerant required is 16 lbs. per cubic foot.
7. For maximum demand, regeneration would be required once a day.

TABLE VI

COST OF DEMINERALIZING RADIOACTIVE WATER
FOR LONG TERM DEMAND

Average Daily Water Requirements in 1000 gallons	Maximum Daily Water Requirements in 1000 gallons	Tanks		Bed		Initial Cost	Operating Cost per 1000 gal.
		No.	Dia. ft.	Depth ft.	Volume ft^3		
100	300	2	4.5	2.52	80	$15,000	$0.15
50	150	2	3	2.83	40	10,000	0.18

Costs will vary somewhat depending on water quality and extent of treatment required for a particular situation, and this can only be evaluated when the problem is at hand. These costs do not reflect the cost of distribution of the water with proper sanitary precautions. At most bases it will probably be more economical to obtain the necessary water from safe ground water sources rather than to resort to this special treatment.

APPENDIX A

SURFACE AND UNKNOWN WATER SOURCES, TREATMENT AND CONSUMPTION FOR U. S. AIR FORCE BASES - FISCAL YEAR 1955*

State	Base	Total Water Use x1000	Amount from Surface Source x1000	Source	Treatment	Remarks
Ala.	Gadsen	10,189	10,189	Coosa R.	Coagulation, Sedimentation Filtration, Disinfection	--
."	Brookley	526,408	663	Big Creek Res.	Coagulation, Sedimentation Filtration, Disinfection	--
Ariz.	Yuma Air-port	314,303	314,313	--	--	Purchased from U.S. Bureau of Reclamation
Calif.	March	598,098	418,154	Colorado R.	Coagulation, Sedimentation Filtration, Disinfection	--
"	Travis	599,592	514,244	Sacramento R.	Coagulation, Sedimentation Filtration, Disinfection	--
"	Norton	399,575	8,109	Lytle Cr. & Devil Canyon	Disinfection	--
"	Beale	401,227	81,000	--	--	Purchased from Nevada Irr. Dist.
"	Hamilton	245,507	245,507	Kent, Alpine, Bon Tempe, Lagunitas	Coagulation, Sedimentation, Disinfection	--
Colo.	Lowry	537,648	537,648	South Platte	Coagulation, Sedimentation, Filtration, Disinfection Softening	Possibly 5 plants

State	Base	Total Water Use x1000	Amount from Surface Source x1000	Source	Treatment	Remarks
Colo.	Ent.	26,641	26,641	Fountain Cr.	Coagulation, Sedimentation, Filtration, Disinfection	--
"	Peterson Field	22,751	22,751	Fountain Cr.	Coagulation, Sedimentation, Filtration, Disinfection	--
Fla.	MacDill	385,138	385,138	Hillsborough River	Softening, Coagulation, Sedimentation, Filtration, Disinfection	--
"	Orlando	180,731	180,731	Lakes Adair, Highland, Spr.	Aeration, Coagulation, Sedimentation, Filtration, Disinfection	--
"	Palm Beach	146,766	12,866	Clear Lake & Mangonia	Aeration, Coagulation, Sedimentation, Filtration, Disinfection.	
Ga.	Dobbins	33,978	33,978	--	--	Purchased from Lockheed A.C., Marietta
Ill.	Scott	469,252	469,252	Miss. R.	Coagulation, Sedimentation, Filtration, Disinfection	--
"	Hanna City	4,769	4,769	--	--	--
"	O'Hare Int'l Airport	105,705	105,668	Lake Michigan	Coagulation, Sedimentation, Filtration, Disinfection	--
Kan.	Forbes	193,155	193,155	Kansas R.	Aeration, Softening, Coagulation, Sedimentation, Filtration, Disinfection	Purchased from Topeka
"	Topeka (AFD)	33,235	33,235	--	--	Purchased from Forbes AFB

State	Base	Total Water Use x1000	Amount from Surface Source x1000	Source	Treatment	Remarks
Ky.	Campbell AFB	--	--	--	--	No data available
La.	Barksdale	482,347	482,347	--	--	--
"	Slack AFD	64,824	64,824	Cross Lake	Aeration, Coagulation, Sedimentation, Filtration, Disinfection	2 Plants
Me.	Dow	178,221	178,221	Penobscot R.	Aeration, Coagulation, Sedimentation, Filtration, Disinfection	--
"	Presque Isle	143,665	143,665	Presque I. Stream	Coagulation, Sedimentation, Filtration, Disinfection.	--
Maryland	Andrews	311,867	309,073	Patuxent R. Anacostia	Aeration, Coagulation, Disinfection	2 Plants
Mass.	Westover	380,822	380,822	Quabbin Res. Coolby Brook	Disinfection	2 Plants
"	L.G.Hanscan Field	101,666	101,661	Lakeville Pond	Disinfection	--
Mich.	Selfridge	193,130	193,130	L.St.Clair	Coagulation, Sedimentation, Filtration, Disinfection	--
Minn.	Duluth Airport	16,471	16,471	Lake Superior	Coagulation, Sedimentation, Filtration, Disinfection.	Purchased from Duluth
"	Minn.St.Paul Airport	46,364	46,364	Miss.R.	Coagulation, Filtration, Disinfection	Purchased from Minneapolis
Mo.	Aero Chart & Info.Ctr.	59,299	59,299	Missouri R.	Coagulation, Sedimentation, Filtration, Disinfection	2 Plants

State	Base	Total Water Use x1000	Amount from Surface Source x1000	Source	Treatment	Remarks
Mo.	Grandview	52,255	52,255	--	Softening, Coagulation, Filtration, Disinfection	Purchased from Kansas City.
Mont.	Mainstrom	155,421	155,421	Missouri R.	Coagulation, Filtration, Disinfection	--
N.H.	Greiner	37,444	37,444	Lake Massabesic	Disinfection	--
N. Y.	Griffiss	196,438	196,000	Fish Creek	Sedimentation, Disinfection	--
"	Sampson	943,410	943,410	Seneca Lake	--	A.F.Treatment Chlorination
"	Stewart	83,911	83,911	N.Y.Water Reservoir	Aeration, Coagulation, Disinfection	
N. C.	Laurinburg	5,150	5,150	--	--	Port. Comm.
"	Pope	99,101	99,101	--	--	Purchased from Fort Bragg
Okla.	Altus	86,725	86,725	--	--	--
"	Tinker	907,484	217	N.Candian R.	Softening, Coagulation, Filtration, Disinfection	--
Oregon	Condon	4,045	83	Supply Unknown	--	--
"	Portland	77,122	77,122	Bull Run R.	Disinfection	--
Pa.	Olmsted	26,268	26,268	--	--	Purchased from Middleton

State	Base	Total Water Use x1000	Amount from Surface Source x1000	Source	Treatment	Remarks
Pa.	Greater Pgh. Airport	23,169	20,777	--	--	Purchased from Moon Township
S.C.	Donaldson	188,800	188,800	Reservoir	Disinfection	--
"	Charleston	123,402	123,402	Goose & Foster Crs. + Edisto R.	Aeration, Coagulation , Sedimentation, Filtration, Disinfection	--
Tenn.	Sewart	272,240	272,240	Stones R.	--	A.F. Treatment
"	Arnold Eng. Dev.Center	261,414	261,414	Elk R.	--	A.F. Treatment
"	McGhee Tyson A/P	28,482	28,482	--	--	Purchased from Alcoa-Typoco
Texas	Bergstrom	233,016	233,016	Colorado R.	Coagulation, Sedimentation, Filtration, Disinfection	--
"	Carswell	340,293	340,293	Lake Worth	Coagulation, Sedimentation, Filtration, Disinfection	--
"	Goodfellow	128,047	128,047	Concho River	Aeration, Coagulation, Sedimentation, Filtration, Disinfection	--
"	Harlingen	237,475	237,475	--	--	--
"	Laredo	112,015	112,015	Rio Grande	Aeration, Coagulation, Sedimentation, Filtration, Disinfection	--
"	Laughlin	167,488	167,488	San Felipe Spring	Disinfection	--

State	Base	Total Water Use x1000	Amount from Surface Source x1000	Source	Treatment	Remarks
Texas	Perrin	115,929	34,372	Red River Schawnee Cr.	Coagulation, Filtration, Disinfection	--
"	Sheppard	591,105	591,105	Lake Kickapoo Wichita R.	Coagulation, Sedimentation, Filtration, Disinfection	--
"	Webb	218,202	218,202	--	Aeration, Coagulation, Sedimentation, Filtration, Disinfection	--
Vt.	Ethan Allen	63,245	912	Lake Champlain	Sedimentation, Filtration, Disinfection	--
Va.	Langley	667,628	667,628	--	--	Purchased from Ft. Monroe
"	Lanley View Gov't.Housing	43,160	43,160	Chickahominy River	Aeration, Coagulation, Filtration, Disinfection	2 Plants
Wash.	Paine	27,851	27,851	Sultan R.	Disinfection	Purchased from Mukitco Everett
Wyo.	F.E.Warren	511,826	511,826	Crystal L.	Aeration, Coagulation, Sedimentation, Filtration	--
D.C.	Bolling	208,270	208,270	Potomac R.	Coagulation, Sedimentation, Filtration, Disinfection	--

State	Base	Total Water Use x1000	Amount from Surface Source x1000	Source	Treatment	Remarks
Hawaii	Hickam	1,308,259	1,308,259	--	--	Purchased from Navy
"	Wheeler	66,416	66,416	--	--	Purchased from Navy
Panama Canal Zone	Albrook	272,247	272,247	--	--	Purchased from Panama Canal Co.
"	France	50,149	50,149	--	--	--
Puerto Rico	Ramey	716,641	716,641	--	--	Purchased from Isabella Irr. Dist.

* Breakdown on all Air Force Supplies:
Air Force owned surface supplies with treatment plants: 3%
Air Force owned wells: 43%
Purchased from municipalities (some wells, some surface): 54%

APPENDIX B

INSTRUMENTATION FOR WATER MONITORING

No specialized equipment for the monitoring of low levels of contamination in water in the presence of high gamma ray background is in existence at the present time. However, discussions between the Corps of Engineers and the Signal Corps have indicated a probe type instrument, which can be immersed into the water under examination, might best be suitable to satisfy the present requirement.

No construction details for such an instrument have been worked out at this time, but it is possible to predict the performance and the limitations of this type of probe from basic considerations. It will probably be desirable to equip this instrument with two detectors, one of which is sensitive only to gamma radiation (from the water under examination and from background radiation); the other sensitive to the aforementioned gamma radiation plus the beta radiation from the water contamination. Special circuitry in the instrument provides for subtraction of the gamma only, from the gamma plus beta, resulting in a beta only indication. The beta background from the "surrounding" or ambient radiation can be neglected because it is completely shielded by the water into which the probe is immersed.

A brief calculation below shows that under these circumstances, it will be possible to measure contamination down to 10^{-4} μc/ml or even 10^{-5} μc/ml if no appreciable outer gamma background is present. A sensitive detector of the Geiger-Mueller or scintillation type should be used, but details are not critical. In the presence of a high gamma outer background, the sensitivity of the detector will be reduced. It is estimated that in an external

field of 1 r/hr, contaminations down to 10^{-2} to 10^{-3} μc/ml will be measureable. In a field of 10 r/hr, contaminations down to 10^{-1} to 10^{-2} μc/ml are possible of measurement.

It is not believed that in the present state of the art a much greater sensitivity can be achieved by using other methods of monitoring at the point of interest. Methods of differentiation between different radioisotopes by purely physical means are so complicated that they need not be considered in this connection. Examination of water samples at rear locations with low radiation background; e.g., by boiling down of the water, will of course lead to much higher sensitivities. All presently known radiac instruments measure only the gross dose rate. Chemical separation methods will have to be used if a detailed determination of special isotopes is proven to be necessary.

SAMPLE CALCULATION

It is assumed that an extended body of water is uniformly contaminated with C microcuries of activity per milliliter of water. The gamma ray dose rate will then be constant throughout the water in all points not too close to the surface and is given by:

(1) $$D.R. = \int_0^\infty \oint \frac{FC}{r^2} e^{-\mu r} \, dr \, d\Omega = 4 \frac{\pi FC}{\mu}$$

D.R. = dose rate in mr/hr.

C = activity concentration in μc/ml.

μ = gamma ray absorption coefficient in cm^{-1}.

F = conversion factor from activity to dose rate.

F depends on the energy of the gamma radiation and the true adsorption coefficient of air at this energy and is of the order of 5 for gamma radiation of 1 mev. Since μ is of the order of 0.04 at this energy,

(2) D. R. = $400\pi C$

which means a dose rate of about 1/6 r/hr for an activity concentration of 1 μc/ml. The beta dose rate which corresponds to this contamination is of the same order of magnitude as the gamma dose rate because the much larger adsorption μ of the betas is compensated by a correspondingly larger specific ionization in the detector (larger F).

If only the natural background is present, dose rates of less than 0.1 mr/hr can be easily detected with sensitive detectors. This means, according to Eq. 2, that contaminations of the order of 10^{-5} μc/ml should be detectable under these circumstances. The existence of strong additional gamma fields introduces the problem of measuring the beta radiation from the contaminated water as the small difference of two large readings. The higher minimum activities given in the text for specified gamma background levels are obtained if a beta dose rate equal to one or a few percent of the gamma dose rate are considered as measurable. Because of the adsorption of gamma radiation of outside origin within the water, the accuracy of the activity determination increases with increasing depth of immersion of the probe.

APPENDIX C

INSTRUMENTATION FOR RADIOACTIVE ASSAY

by
William K. Muschler, Assistant Sanitary Engineer

EQUIPMENT

Three basic types of radioactive detecting devices were used during this project. The individual, potentially hazardous, beta-emitting radionuclides were quanitatively measured by use of internal proportional counters as manufactured by Nuclear Measurements Corporation. This equipment included either a PCC-10 or PCC-11 Proportional Counter Converter and a Scaler of either the binary or decade type. This type of instrument was employed because of its high efficiency for detecting low levels of activity.

For the higher activity beta samples an Automatic Sample Changer manufactured by Tracerlab, Inc. was used. This unit consists of a SC-5l Autoscaler, a SC-6D Automatic Sample Changer, a SC-6E Shielded Manual Sample Changer and a SC-5F Tracergraph Printer. Because of the continuous, unattended operation of this system, it was employed for decay studies on the higher activity beta samples, specifically the gross beta samples.

In order to obtain qualitative data on the success of the various radiochemical procedures, a Twenty Channel Differential Pulse Height Analyser, Model 520 Modified, manufactured by Atomic Instrument Company was employed. This machine together with a lead shielded sodium iodide (thallium activated) well crystal made it possible to scan the major

portion of the gamma energy spectrum and locate qualitatively the gamma emitting radionuclides present. In addition, where reference sources of specific nuclides were available, a quanitative measure of their activity was possible.

INTERNAL PROPORTIONAL COUNTING

Because of some revision in the sample preparation during the period of investigation, it was necessary to count two physically different types of samples in the internal proportional counters. Initially, all samples were prepared on thin, 2-inch, pressed-aluminum planchets with radioactive material placed directly in planchets. As the techniques of radiochemical separation were improved, samples were prepared on 7/8-inch diameter filter paper and placed in rigid 1-inch stainless steel planchets.

The procedure for use of internal proportional counters are as follows:

1. Each sample would be counted for two equal periods of time.
2. Because of the number of samples, no samples would be counted for more than two twenty-minute counts.
3. When possible, samples would be counted for a period of time to yield a count having a percent error at the 95 percent confidence level of less than 3 percent.

The normal operating day begins and ends with a two-minute count on a source of radioactivity of extremely long half-life and a twenty-minute count of background. The source used was uranium oxide placed in a planchet and covered with one sheet of aluminum foil. The purpose of

the foil is to exclude all alpha particles from the detector. By making a count of the uranium oxide source, any major defects in the counter can be discovered and remedied. The background count is made with an empty planchet placed in the counting chamber. By making a twenty-minute count at the beginning and end of the counting day, an average background count rate can be subtracted from sample count rate of all samples run regardless of length of counting time.

The length of counting time is dependent on the amount of radioactivity in the sample. Based on the previously mentioned rules for internal proportional counting, an error of less than 3 percent at the 95 percent confidence level was the goal. Naturally, there were some samples which did not have sufficient activity to have an error of less than 3 percent within the maximum allowable time. These are treated on an individual basis to determine actual error at the 95 percent confidence level. For the operation the following counting times are set based on computation of 3 percent error:

Gross Counts per minute	Counting Time (in minutes)
less than 300	2 x 20
greater than 300, less than 600	2 x 10
greater than 600, less than 1200	2 x 5
greater than 1200	2 x 3

For decay studies, frequency of counting a particular sample is dependent on the isotope or isotopes present. Generally, it is sufficient to secure three readings per half-life of the isotope. In some cases, where the isotope or isotopes are not well defined, an arbitrary decision is necessary. For mixtures of isotopes one reading a week should suffice. The gross beta samples are counted once every ten days.

END-WINDOW COUNTING

The types of samples counted on the automatic sample changer were mainly gross beta samples. The overall efficiency of the end-window counter ranges from below 10% to 18% depending on the thickness and energy range of the sample. This is about one third of the efficiency of the internal proportional counters. Because of this low efficiency of the end-window counter, only the more active samples could be counted. The samples were limited, by the apparatus, to a maximum diameter of one inch. Twenty-five samples could be accomodated at any one time.

The operation of the automatic sample changer is based on a preset count. For this reason the counting procedures differ from those employed on the internal proportional counters. Since the machine functions continuously, the normal operating procedure for the automatic sample changer is as follows:

Depending on the length of count and the activity of the samples, a minimum of two and a maximum of five background (empty) planchets are interspersed between the samples. The desired result is at least two

background counts per day and not more than five samples between two background planchets.

During this project the changer was so controlled that approximately one count was made on each sample per day. Since the results are automatically printed, the operation of the changer can be checked by simply observing the values of any sample over a period of days.

SCINTILLATION COUNTING

Initially a single-channel pulse height analyser was used for the detection of gamma emitting isotopes. In September, 1957, a 20-channel differential pulse height analyser was made available. This made the qualitative scanning of the sample possible. In scanning a sample of mixed radioisotopes, at best, only a qualitative result was possible. When a source of known activity was available and that specific nuclide was under investigation, a reasonable estimate of the activity of the nuclide could be made.

The general procedure followed on operation of the 20-channel differential pulse height analyser was to run a test pulse through each of the twenty channels to check the window widths. After sufficient counts were registered to produce a statistically valid result, the widths were adjusted to produce an error of less than 2 percent.

The analyser was then adjusted so that the known energy peak of a reference source coincided with the proper channel of the analyser. The reference source used was Cs^{137} having an energy peak at 0.662 million electron volts.

The majority of the samples scanned had sufficient activity to reduce the counting error to less than five percent in the maximum counting time of 32 minutes for any settling of the twenty channels. A background count was taken during the middle of the operating day and subtracted from the gross scans. The mixed isotopes samples were counted once a month over the spectrum from 0.10 to 1.4 million electron volts. Since the information obtained was only qualitative, the procedure proved adequate.

CORRECTION FACTORS

In order to convert the net count rate obtained on the internal proportional counters and end-window counters into absolute activity, it was necessary to obtain calibrated sources. Plated calibrated sources were not applicable because of the nature of the sample and sources that had to be made up. The efficiency of any counting system is dependent on the geometry of the system, the backscatter of the materials used, the self-absorption of the sample, the energy of isotopes involved and windows and air losses in the end-window counters. It is possible to theoretically adjust for each of these factors, however, it was decided to lump all these adjustments together and work with only one set of curves. If the same detectors and materials are used in the preparation and counting of sources and samples, it is then possible to plot a single curve of efficiency versus thickness of sample for a particular isotope.

The initial samples plated in 2-inch aluminum dishes were converted to absolute activity by using the correction factors previously determined

by the Radiological Health Program at the Sanitary Engineering Center. When the radiochemical techniques were altered to produce samples in 1 inch stainless steel planchets, it became necessary to produce sources of approximately the same physical characteristics as the samples. The procedure was to plate out a small quantity of isotopes on thick flat copper planchets in triplicate. Since the geometry of the internal proportional counter is 0.50 for a flat planchet and the thickness of the sample was approximately zero, the average of the count rate could be converted by means of backscatter curves[1] to absolute activity. With the activity of the isotope known, five to ten samples were prepared with a constant amount of activity and varying amounts of carrier added. By comparing the sample count rate to the disintegration rate added to the sample an overall efficiency was determined. This procedure was followed with Ca^{45}, Sr^{89}, Y^{91}, Cs^{137}, and Tl^{204} with varying degrees of success. Adequate curves were obtained for Ca^{45}, Sr^{89}, and Tl^{204} and were used for analysis.

Since radioactive disintegration is a random occurrence, there is attached to any measurement of it a statistical error. For this project the 95 percent confidence level was chosen and the error of the net counting rate was determined by the following formula:

$$E_{95} = 1.95 \left(\frac{N_s}{T_s} + \frac{N_B}{T_B} \right)^{1/2}$$

N_s = Gross counting rate

N_B = Background counting rate

T_s = Total sample counting time

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(1) "Evaluating the Performance of the Internal Counter" by J. R. Nelson, G. R. Hagee, C. R. Setter. Nucleonics, Vol. 12, No. 6, June, 1954.

APPENDIX D

by

Herman L. Krieger, Chemist

ANALYTICAL METHODS

A step-wise procedure that was followed to ascertain the total strontium activity in the Nevada Test Site soil samples is tabulated below:

1. In a 50-ml centrifuge tube weigh out 1 to 5 grams of soil sample.
2. Leach soil with 6M HNO_3, adding four consecutive 25-ml portions, stir and heat thoroughly after adding each. This will remove about 75 to 80 percent of the total activity involved.
3. The strontium procedure follows:
   a. To an aliquot of the leach add 20 mg of Sr^{++} carrier (weighed as the oxalate) and 20 mg of Ba^{++} carrier (weighed as the chromate.) Stir and heat to a boil. Make basic with 19 M NaOH, add 5 ml 2M Na_2CO_3 and stir in ice bath until cool. Centrifuge and discard supernate.
   b. Wash the precipitate with 25 ml of distilled water.
   c. Dissolve the precipitate with 5 ml of 6 M HNO_3, place in ice bath. Add 25 ml of fuming HNO_3 and stir to precipitate $Sr(NO_3)_2$. Centrifuge and discard supernate into a beaker of water. Let tube drain inverted for several minutes.

d. Wash precipitate twice with 25 ml of acetone. Drain tube thoroughly.

e. Dissolve the precipitate in 5 ml of distilled water, place in ice bath. Add 20 ml of fuming HNO_3 and stir to precipitate $Sr(NO_3)_2$ again. Centrifuge and discard supernate into a beaker of water. Drain inverted tube for several minutes.

f. Dissolve the precipitate in 10 ml of distilled water, add 5 mg Fe^{+++} and a small amount of NH_4OH to produce a precipitate of $Fe(OH)_3$. Stir, centrifuge and transfer supernate to a clean centrifuge tube. Discard $Fe(OH)_3$ precipitate.

g. Adjust the supernate to neutral using phenolphthalein indicator and 1 M HNO_3. Buffer the sample with 1 ml 6 M HAc and 2 ml of 6 M NH_4Ac. Bring to a boil and slowly add 1 ml 1.5 M Na_2CrO_4 to precipitate $BaCrO_4$. Cool, stir, and centrifuge. Transfer the supernate to a clean centrifuge tube. Discard (or save for Ba analysis) the chromate precipitate.

h. Add 2 ml NH_4OH to the supernate and bring to a boil Add 5 ml saturated $(NH_4)_2C_2O_4$ and place in ice bath. Stir until cool. Centrifuge and discard supernate.

i. Take up precipitate in 10 ml hot water to dissolve excess oxalate. Filter on tared paper. Wash with hot water, C_2H_5OH and dry with ether.

j. Count

Other radionuclides were analyzed with previously developed methods that appear in the literature.